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## LEAD SLOWING-DOWN SPECTROMETER RESEARCH AT LANSCE\*

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The lead slowing-down spectrometer (LSDS) at Los Alamos is a 20 ton cube of lead with numerous channels, one for the proton beam from the LANSCE accelerator and others for samples and detectors. A pulsed spallation neutron source at the center of the cube is produced by the 800 MeV proton beam incident on an air-cooled tungsten target. Neutrons from this source are quickly downscattered by various reactions until their energies are less than the first excited state of  $^{207}\text{Pb}$  (0.57 MeV). After that, the neutrons slow down by elastic scattering where they lose on the average 1% of their energy per collision. The mean energy of the neutron distribution then changes with time as  $\langle E \rangle \sim 1/(t + t_0)^2$ , where " $t_0$ ," is a constant. The low neutron absorption cross section of lead and multiple scattering of the neutrons leads to a very large neutron flux, approximately 1000 times that available in beams at the intense neutron source at the Lujan Center at LANSCE. Thus nuclear cross sections can be measured with very small samples, or conversely, very small cross sections can be measured with somewhat larger samples. Present research with the LSDS at LANSCE includes measuring fission cross sections on short-lived isotopes such as  $^{237}\text{U}$ , developing techniques to measure (n,p) and (n,alpha) cross sections, testing new types of detectors for use in the extreme radiation environment, and, in an applied context, assessing the possibility of measuring the isotopic content of actinide samples with the eventual goal of characterizing fresh and used reactor fuel rods.

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## 1. Introduction

Lead Slowing-Down Spectrometers (LSDS) are specialized tools in the experimenters' tool box for neutron science and engineering. The concept of an LSDS was originated by Shapiro and Bergman in 1955 [1,2], and since then, several have been constructed. Although each spectrometer fielded so far is unique in its size, shape and driving neutron source, they all operate on the same principle. A pulsed neutron source injects neutrons into a large mass of pure lead, where they inelastically scatter or make reactions until their energies are less than the first inelastic level in stable lead isotopes (0.570 MeV in  $^{207}\text{Pb}$ ). Then they slow down by elastic scattering, where they lose on the average 1% of their energy per scattering. Because lead has a very low cross section for capturing neutrons, the neutrons continue to scatter until they escape from the lead or reach low energies. With a large mass of lead with a small surface-to-volume ratio, the leakage is small and so the neutrons are, in a sense, trapped by the lead. To use this large neutron flux, samples and detector are put inside the lead volume, where the neutron flux in the range of 0.1 eV to over 100 keV is larger than that offered by traditional beam-target setups at modern accelerator facilities by two to three orders of magnitude. Thus many measurements that were very difficult or impossible by conventional beam-target setups have been made possible by the LSDS.

This report focuses on the LSDS at the Los Alamos Neutron Science Center (LANSCE) where the driving neutrons are produced by a spallation neutron source of 800 MeV protons on a tungsten target. This is classified by Alexeev et al. [3] as a "third generation" LSDS as suggested by Stavitsky [4] and Moore [5]. The first generation is driven by 14 MeV D-T neutron generators or Van de Graaff accelerators with a variety of neutron-producing reactions. The second generation LSDS is driven by an electron linear accelerator to produce neutrons by photonuclear reactions. And the third generation is driven by spallation neutron sources. The generations are ordered by the intensity of the driving neutrons. Because spallation reactions produce more neutrons per second per unit beam power and because the limiting factor in beam power is dissipation of heat from the target, the third generation LSDS generally offers a greater neutron flux for experiments. The neutron flux on a sample in the LSDS is compared with what is available at LANSCE for beam-target experiments at the Lujan Center with moderated neutrons and at the unmoderated Weapons Neutron Research (WNR) source in Figure 1. The much greater flux with the LSDS enables experiments that are impossible or impractical at other facilities.

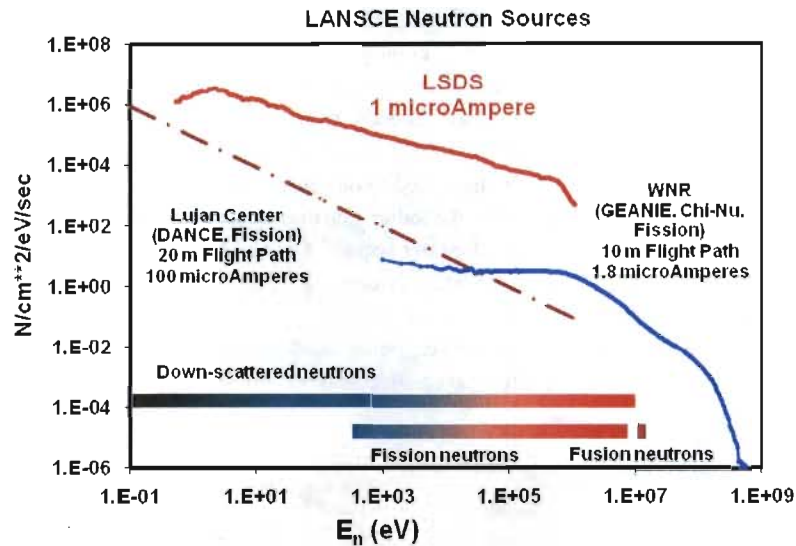


Figure 1. Neutron flux available as a function of neutron energy for three sources at LANSCE. The typical time-averaged beam current is shown as well as typical flight path lengths for the Lujan and WNR facilities. Regions of interest for energy applications (fission, fusion, etc.) are also shown.

## 2. The LANSCE LSDS

The LSDS at LANSCE [6,7] consists of an assembly of 36 lead blocks, each  $40 \times 40 \times 30 \text{ cm}^3$  stacked to form a cube 1.2 meters on a side. It is covered with 0.7 mm of cadmium to prevent neutrons that escape the lead and are thermalized in the room from returning to the spectrometer. This assembly was designed and fabricated in France to the specifications of researchers at CEA in Bruyères-le-Châtel where it was used with a neutron source produced by a Van de Graaff accelerator [8]. Many of the blocks have channels, 10 cm wide by 5 cm high, which can be used to insert samples and detectors. Two such channels are combined to produce a 10 cm x 10 cm channel that extends through the assembly where a beam from an accelerator can strike a target in the middle of the lead.

For use at LANSCE, a tungsten target, 25 cm long by 7 cm in diameter is placed in the middle of the lead. It is cooled by forced air. A pulsed 800-MeV proton beam from the accelerator is directed to this target. For low intensity experiments, the beam can be used directly from the linear accelerator. A stream of micropulses, each having  $3 \times 10^8$  protons, spaced 5 ns apart, comprises the incident pulsed beam. The pulses need to be separated by many milliseconds to avoid pulse overlap, and with the other constraints of the accelerator, the pulse rate is typically up to 40 pulses per second. For larger beam pulses, or for shorter incident pulse widths, the beam is compressed in a proton storage ring [9,10] before being directed to the target. Typical beam pulses are less than 100 ns long. The maximum time-averaged beam used at present is limited to 1 microampere for radiation protection considerations.

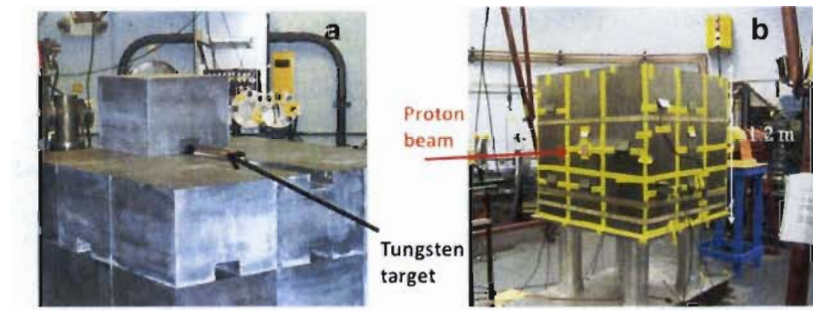


Figure 2. The LSDS at LANSCE: (a) Partially assembled and (b) fully assembled and covered with 0.7 mm of cadmium.

The energy distribution of the neutron flux has been modeled by Monte Carlo calculations as shown in Figure 3 [7]. The neutrons initially have a very wide distribution in energy. After about 1 microsecond the distribution narrows and attains a width of 30% in  $DE/E$  after about 10 microseconds. The mean energy of the distribution is given as  $\langle E \rangle = K / (t + t_0)^2$  where the constants were determined experimentally to be  $K = 161 \pm 1 \text{ keV} \cdot \mu\text{s}^2$  and  $t_0 = 0.4 \pm 0.1 \mu\text{s}$ .

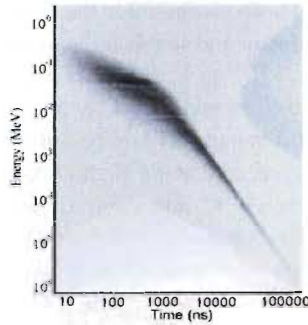


Figure 3. Monte Carlo simulation of the neutron energy distribution in the LSDS as a function of time. After a microsecond or so, the distribution narrows and attains a width of about 30% in  $\Delta E/E$ . The figure is from [7].

### 3. Uses of the LANSCE LSDS

This section describes the uses to which we are putting the LSDS at LANSCE. They include fission cross section measurements, (n,p) and (n,alpha) cross section measurements and development of techniques for using the spectrometer to assay the amount of fissile isotopes in reactor fuel, both fresh and irradiated.

#### 3.1. Fission cross section measurements

The initial goal for installing the LSDS at Los Alamos was to measure neutron-induced fission cross sections on isotopes that had short half lives or were otherwise very difficult to obtain or handle. One of these isotopes is in fact the isomer of  $^{235}\text{U}$ , which lies at only 76 eV above the ground state and has a half life of 26 minutes. This isomer is populated nearly 100% from the alpha decay of  $^{239}\text{Pu}$ , and therefore there is the possibility of obtaining the isomer by extracting uranium chemically from  $^{239}\text{Pu}$ . Separations of a small amount of the isomer made possible several measurements of the fission cross section of this isotope for thermal and cold neutrons but not for epithermal neutrons. Several calculations indicated that the fission cross section of this isomer should be similar to that of the ground state but, depending on the incident neutron energy, as much as 40% lower. The LSDS experiments could also elucidate the fission resonance structure for a completely different range of spins of the compound nucleus, as the ground state spin-parity of  $^{235}\text{U}$  is  $7/2^-$  whereas that of the isomer is  $1/2^+$ . For the LSDS measurement, it was calculated that 10 – 20 ng of the isomer would be required, a much larger amount than was used in the measurements at thermal because the cross section was higher than that expected for epithermal neutrons by an order of magnitude or more. The

separation procedure that had been successful in the smaller amounts turned out to fail when scaled up by this factor and so this measurement was not successful.

Another isotope whose fission cross section has not been well measured is  $^{237}\text{U}$  (6.7 day half life), and a project is underway to measure its fission cross section with the LSDS. The sample is prepared at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory by subjecting a highly enriched sample of  $^{236}\text{U}$  to thermal neutrons. The isotope is then fashioned into a compensated fission ion counter that uses the compensation technique of Koehler et al. [11] and that is pictured schematically in Figure 4. The  $^{237}\text{U}$  is deposited on one side of the center electrode and a reference sample of  $^{235}\text{U}$  is deposited on the other. A typical signal, see the figure, consists of positive pulses from  $^{237}\text{U}$  and negative ones from  $^{235}\text{U}$ . Thus the fission cross section of  $^{237}\text{U}$  can be measured relative to the well known fission cross section of  $^{235}\text{U}$ . This experiment is underway now and results should be forthcoming within a year.

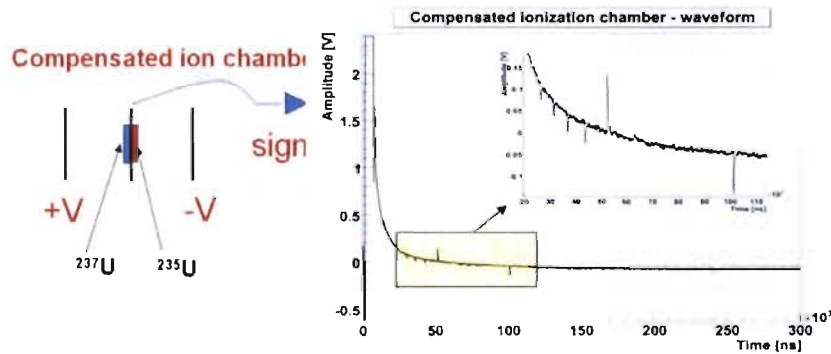


Figure 4. Schematic of the compensated ion chamber used in the measurement of the fission cross section of  $^{237}\text{U}$  and the signal which contains fissions of  $^{237}\text{U}$  (positive going short pulses) and fissions of  $^{235}\text{U}$  (negative going pulses). The smooth decrease in the base line is from recovery of the preamplifier following the large "gamma-flash" at time zero.

### 3.2. $(n,p)$ and $(n,\alpha)$ cross section measurements

Low energy neutron reactions with complex nuclei do not often result in the emission of light charged particles such as protons and alpha particles. For most stable nuclei, the  $(n,p)$  reaction has a negative  $Q$ -value and is therefore not allowed. Unstable nuclei, however, especially those that are proton rich, are more likely to have positive  $Q$ -values for  $(n,p)$  reactions. These reactions are of

interest for basic physics of reactions on unstable nuclei and in designing transmutation process to reduce radioactive waste. The (n,α) reaction has a positive Q-value for many isotopes, but the cross sections for this reaction are often small because the alpha particles need to pass through the Coulomb barrier and those transmission coefficients can be small. Yet both of these reactions are of interest to a better understanding of nucleosynthesis in astrophysics and to the tests of basic reaction physics models.

Experimental study of these reactions requires the detection of particles that have energies an order of magnitude smaller than those of fission fragments of the previous section. Thus, for example, the peaks in Figure 4 above will be that much smaller. If charged particle spectroscopy is desired to learn which states in the residual nucleus are reached, then the sample needs to be very thin to allow the charged particles to escape with little energy loss. Thin targets and small cross sections present a challenge to the experimenter.

Several years ago, we used an ion chamber to demonstrate that (n,α) reactions could be studied with the LSDS by measuring this cross section on  $^6\text{Li}$ , where the cross section is large [12]. More recently, we are using Passivated Implanted Planar Silicon (PIPS) silicon detectors, which were developed and tested for LSDS use at Rensselaer Polytechnic Institute [13]. They seem to have a reasonable lifetime in the extreme radiation environment of the LSDS. Again, a variation of the compensation technique is used by placing a second detector, which does not see the sample, close to the first and then do a subtraction of the two signals before introducing the net signal to the preamplifier. The present concept for housing the two detectors is shown in Figure 5. Studies of (n,p) and (n,α) reactions on  $^{50}\text{V}$ ,  $^{147}\text{Sm}$  and  $^{149}\text{Sm}$  is in progress.

### ***3.3. Assay of fissile isotopes in reactor fuel***

The LSDS has for many years been considered as a tool to assay the content of fissile isotopes in nuclear fuel [14]. The idea is to use the time-dependent energy spectrum of the spectrometer to sweep through fission resonance of the fissile isotopes in the sample. Detection of fissions can be accomplished by looking at the fast fission neutrons, which have energies of 1 MeV and higher, much higher than those inducing fission in the resonance region. Because every isotope has its own "signature," the amount of each will be revealed by the time history of the fast fission neutrons. The challenge then is to develop a detector



that detects the MeV neutrons but, at the same time, is “blind” to the eV and keV neutrons inducing the fission.

We have used detectors that contain  $^{232}\text{Th}$  or highly depleted uranium that is essentially pure  $^{238}\text{U}$ . Both of these isotopes have thresholds for fission that are in the 100's of keV, and therefore they are not fissionable by neutrons below this energy. To increase the detection sensitivity of a fission counter, we have developed a multiplate fission with  $^{232}\text{Th}$  as the active material (see Figure 6). Again, the technique of compensation is used. An example of the data obtained is given in Figure 7 where a sample of  $^{235}\text{U}$  was placed near this fission counter.



Figure 5. Concept of having two PIPS detectors in the same housing. The sample is on a backing (i.e. the blue region on the center disk) so that the protons and alpha particles can be detected by only one detector.

### 3.4. Further detector development

From this discussion, one can see how important it is to develop novel detectors for use in the LANSCE LSDS. We are working on diamond detectors, both single crystal and polycrystalline, which have remarkable radiation resistance. They have the speed and stopping power to be attractive for (n,p) and (n,alpha) reaction studies. Other detectors under consideration are Gas Electron Multipliers (GEM), solar cells, and silicon carbide detectors.

## 4. Summary

The LANSCE LSDS provides a source of neutrons in the energy range 0.1 eV to 100 keV that is two to three orders of magnitude more intense than that available in neutron beams at LANSCE, which is one of the most powerful spallation neutron sources in the world. A wide range of new experiments is therefore possible with the LSDS.

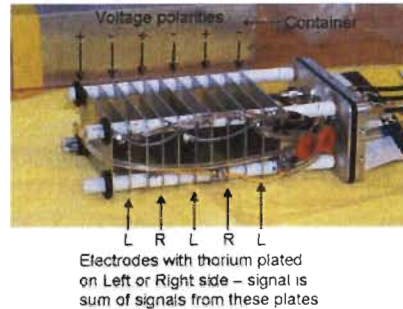


Figure 6. Multi-plate fission ion chamber containing  $^{232}\text{Th}$ . The electrode arrangement is designed to compensate the large signal from the gamma-flash before the signals are sent to the preamplifier.

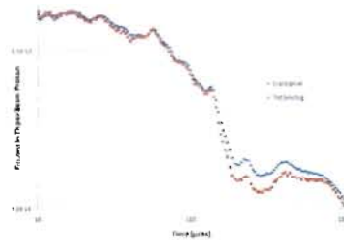


Figure 7. Time history of fast neutrons from fission of  $^{235}\text{U}$  in the resonance region. The shape of the data indicates that the only fissile isotope in the sample was  $^{235}\text{U}$ .

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### References

1. L. E. Lazareva, E. L. Feinberg, and F. L. Shapiro, *Zh. Eksp. Teor. Fiz.* **29**, 381 (1955).
2. A. A. Bergman, A. A. Isakov, A.I. Murin, F. L. Shapiro, I. V. Shtranikh, and M. V. Kazarnovsky, *Proc. Int. Conf. Peaceful Uses of Atomic Energy, Geneva, 1955*, vol 4, 166 (1955).
3. A. A. Alexeev, Yu. V. Belousov, A. A. Bergman, A. N. Volkov, Yu. M. Gledenov, O. N. Goncharenko, M.N. Grachev, M. V. Kazarnovsky, V. L. Matushko, V. L. Matushko, V. I. Mostovoy, A. V. Novikov, S. A. Novoselov, S. S. Parzhitski, Yu. P. Popov, Yu. A. Ryabov, and Yu. Ya. Stavitsky, "Third Generation of Lead Slowing-Down Spectrometers: First Results and Prospects," *Phys. At. Nuclei* **62**, No. 5 793 (1999); trans *Yad. Fiz* **62**, No 5, 851 (1999).
4. Yu. Ya. Stavitsky, *Yadrena Energiya* **25**, 3 (1987).
5. M. S. Moore, P. E. Koehler, A. Michaudon, A. Schelberg, Y. Danon, R. C. Block, R. E. Slovacek, R. W. Hoff, and R. W. Loughheed, *Proc. Symp. Capture Gamma-Ray Spectroscopy*, ed. R. W. Hoff, AIP Conf. Proc. **238**, 953 (1991).
6. T. Granier, L. Pangault, T. Ethvignot, R. C. Haight, X. Ledoux, V. Méot, Y. Patin, P. Pras, M. Szmigiel, R. S. Rundberg and J. B. Wilhelmy, *Nucl. Instr. Meth.* **A506**, 149 (2003).
7. D. Rochman, R. C. Haight, J. M. O'Donnell, A. Michaudon, S. A. Wender, D. J. Vieira, E. M. Bond, T. A. Bredeweg, A. Kronenberg, J. B. Wilhelmy, T. Ethvignot, T. Granier, M. Petit, and Y. Danon, *Nucl. Instr. Meth. in Phys. Res.* **A550**, 397 (2005).
8. X. Ledoux J. Sigaud, T. Granier, J-P. Lochard, Y. Patin, P. Pras, C. Varignon, J-M. Laborie, Y. Boulin, and F. Gunsing, *Eur. J. Phys.* **A27**,59 (2006).
9. P. W. Lisowski, C. D. Bowman, G. J. Russell, and S. A. Wender, *Nucl. Sci. Eng.* **106**, 208 (1990).
10. P. W. Lisowski and K. F. Schoenberg, *Nucl. Instr. Meth. Phys. Res.* **A562**, 910 (2006).
11. P.E. Koehler, J.A. Harvey, N.W. Hill, *Nucl. Instr. and Meth.* **A361**, 270 (1995).
12. D. Rochman, R.C. Haight, J.M. O'Donnell, S.A. Wender, D.J. Vieira, E.M. Bond, T.A. Bredeweg, J.B. Wilhelmy, T. Granier, T. Ethvignot, M. Petit, Y. Danon, C. Romano, *Nucl. Instr. Meth. Phys. Res.* **A564**, 400 (2006) .
13. Y. Danon, R. Block, J. Thompson and C. Romano, *J. Korean Phys. Soc.* **59**, No. 2, 1649 (2011).
14. Abdurrahman, N.M., Block, R.C., Harris, D.R., Slovacek, R.E., Lee, Y-D. and Rodriguez-Vera, F., "Spent-Fuel Assay Performance and Monte Carlo Analysis of the Rensselaer Slowing-Down-Time Spectrometer," *Nucl. Sci. Eng.* **115**, 279 (1993).